MAGNESIUM ION SOURCE FOR HIGH INTENSITY MASS SPECTROGRAPH

Thesis by

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SUMMARY.

Magnesium ions were produced by bombarding magnesium vapor by electrons emitted from oxide coated cathodes. The ions were then focussed into a thin sheet by means of an electrostatic lens. This ion sheet was sent into the magnetic lens which focussed the sheet into a small spot where they were collected. The ion current obtained at the collector was 120 microamperes.

INTRODUCTION.

Mass spectrographs can, for the present discussion, be divided into two classes: (a) those intended for accurate mass and abundance ratio measurements and (b) those that are built for specimen collection. For class (a), high resolution and strict collimation are essential. The number of ions required need not be very large. "Point" sources are generally used. When the intensity of the ion current is too high, the beam will spread because of the space charge effect, and it is much more serious than in the case of electrons which for the same energy have much larger velocity. When it comes to collecting a specimen of, say, one milligram, the high intensity is necessary. The resolution need not be too high. High resolution and high intensity are incompatible. If again a "point" source is used, there is a definite upper limit for the ion beam current beyond which no sharp beam can be obtained. Tuve [1] and his collaborators had shown experimentally that there is considerable spreading in projecting ion beams of much more than 10 microamperes to distances much in excess of 20 centimeters.

In 1929, W. R. Smythe and L. H. Rumbaugh [2] designed and built a magnetic lens which is capable of focussing a sheet of ions of the same e/m to a point. Since the ions are spread out into a sheet except close to the focal point, the total beam current can greatly be increased. The ion

source used was Kunsman catalyst which was available for a few elements. J. Evvard [3] tried the electron bombardment method for magnesium. The electron emitter he used was a tantalum strip of dimensions 0.005" X 0.085" X 10". The two strips in parallel took about 40 amperes at 40 volts to get an emission of 100 milliamperes. He was able to get an ion current at the collector of 24 microamperes with good resolution. It seems that there are still possibilities of greatly increasing the collector current. This is the main object of the present work.

In order to get large ion current at the collector, five requirements must be satisfied. (1) There must be plenty of magnesium vapor in the ionization chamber. (2) There must be enough electrons to bombard the vapor with. (3) The field just above the slit of the chamber must be strong in order to draw out all the ions that are pushed out of the slit. (4) The lens must be able to focuss them into a thin sheet. (5) This ion sheet must be parallel and directly beneath the pole pieces so that they all can go into the magnetic lens.

The first requirement was met without any difficulty while the fifth one chiefly concerns the alignment which is the more critical the sharper the ion beam is. An oxide coated cathode supplies ample electron emission which meets the requirement(2). The third and fourth ones depend upon the structure of the electrostatic lens. These will be discussed in detail later.

Tantalum was not a very efficient emitter. It took 1.5 kw power to produce 100 ma electron current. Even though there was a water cooling jacket, the soapstone insulators which were rather close to the tantalum filaments sometimes got hot enough to be conducting. The first thing was to find another more efficient emitter. Oxide coated cathodes were then tried without much success at first. When the anode voltage was first applied, the emission was about 40 ma, but very soon, it dropped practically to zero. It was

found that the cathode turned into gray instead of white color. This contamination was due to the gas given out when the magnesium heater was heated. If the magnesium was heated first and then the cathode activated, it stayed active.

Then it was thought that high frequency voltage across two cathodes would accelerate the electrons so that they could ionize the magnesium vapor. Since the ions are so much heavier, they should not be influenced by the high frequency field if the frequency is high enough. The arrangement was as shown in Fig. 1.

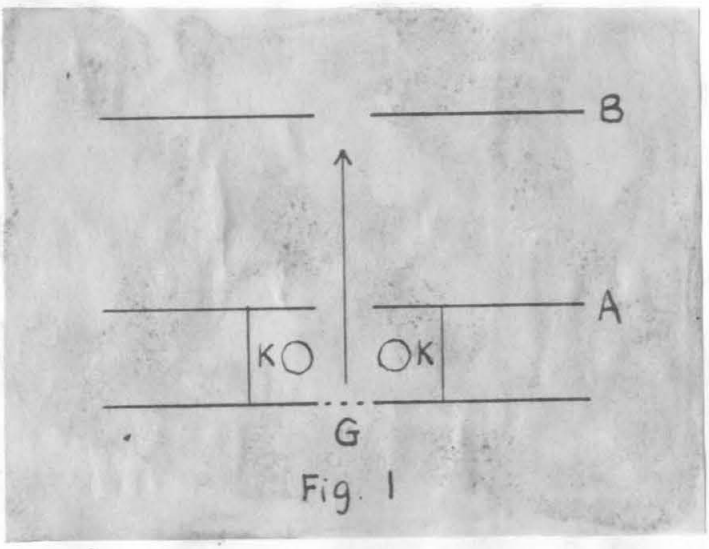


Fig. 1

G was a grid made of copper wire. D.c. potential was applied between cathodes K and G making G positive with respect to K. G was also connected to the positive terminal of the 6000 V d.c. generator. B was grounded. The potential of A could be changed continuously by means of a potentiometer arrangement

so as to obtain the best focussing condition. Without a high frequency field between the two cathodes, a collector current of 7 microamperes was obtained. About the same current could also be obtained with 100 volts high frequency voltage alone. When both were used, the collector current was always smaller. The main disadvantage of this arrangement was that the focussing field greatly affected the field which was responsible for ion production. Another source was built where the main feature was just to isolate this interaction between the focussing field and the field for the ion production. This was the final form adopted. The description and dimensions are given in the following sections.

The first lens tried with the above ionization chamber consisted of three parallel plates each with a slit in the middle of 5 mm width (Fig. 2). With 130 ma cathode

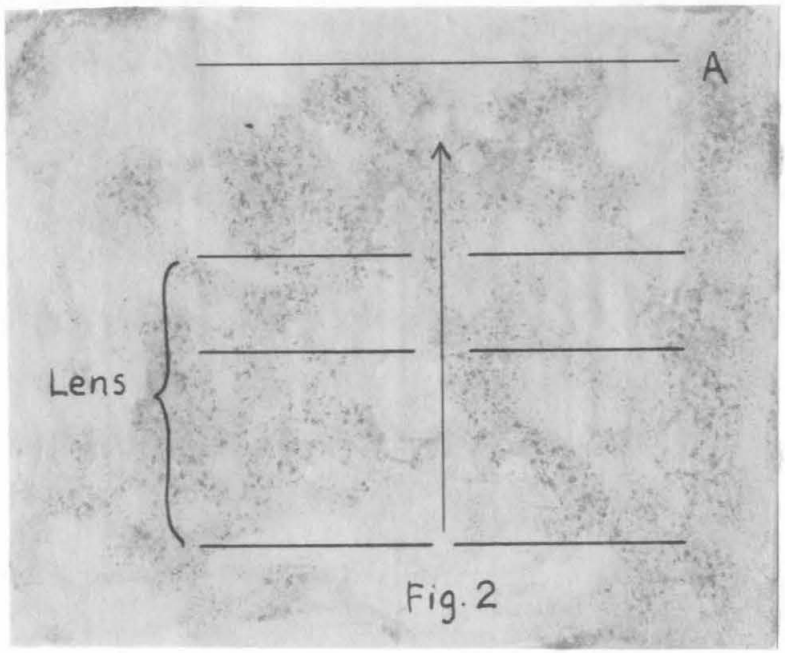


Fig. 2

emission and proper focussing, the ion current to A was 320 microamperes. A very sharp image was found on A which was 2 centimeters above the top plate of the lens. Evidently the lens was too strong. The spacing between the three plates was changed to that shown in Fig. 3. With this lens,

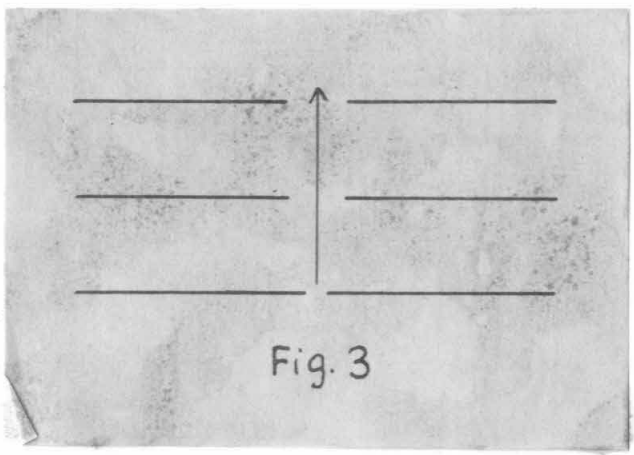


Fig. 3

the largest ion current at the collector was 60 microamperes which was far too small in comparison with the total beam current from the slit. This meant that the beam was still divergent so that only the small central part can get through while about four fifth of them hit the pole pieces.

The focal length of the lens could be calculated but the calculations of the potential distribution were a little involved and there is another kind of lens which will give a stronger field near the ionization chamber slit. So this was abandoned.

In order to test whether this small collector current was due to the defects in the focussing or the defects in the alignment of the different parts, an arrangement like that in

Fig. 4 was set up. Two deflecting plates D_1 and D_2 were installed just on top of the lens and a deflecting voltage was applied. Another pair of plates P_1 and P_2 were put in just below the magnetic pole pieces. The distance between P_1 and P_2 was equal to the gap between the pole pieces which was 6 mm. So if the beam was wider than that, they both sould receive ions perhaps not of the same amount if the beam was off to one side.

The results of some tests showed that P_1 and P_2 received about the same ion currents which fact proved that the alignment was good and that the beam was too broad for the gap between the pole pieces. When a large deflecting voltage was applied between D_1 and D_2, the beam could be directed to either P_1 or P_2 which amounted to about 150 microamperes. Therefore the whole problem was at this stage reduced to an electron optical one.

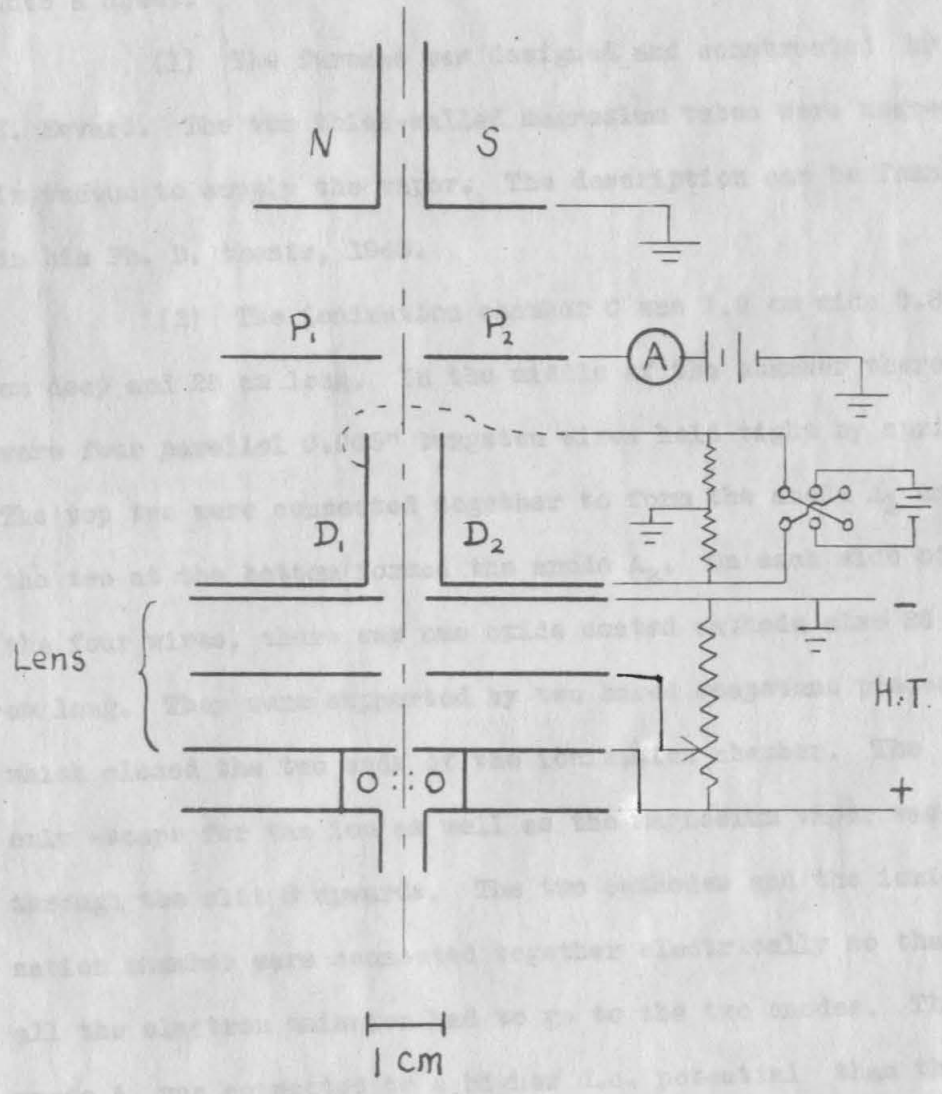


Fig. 4

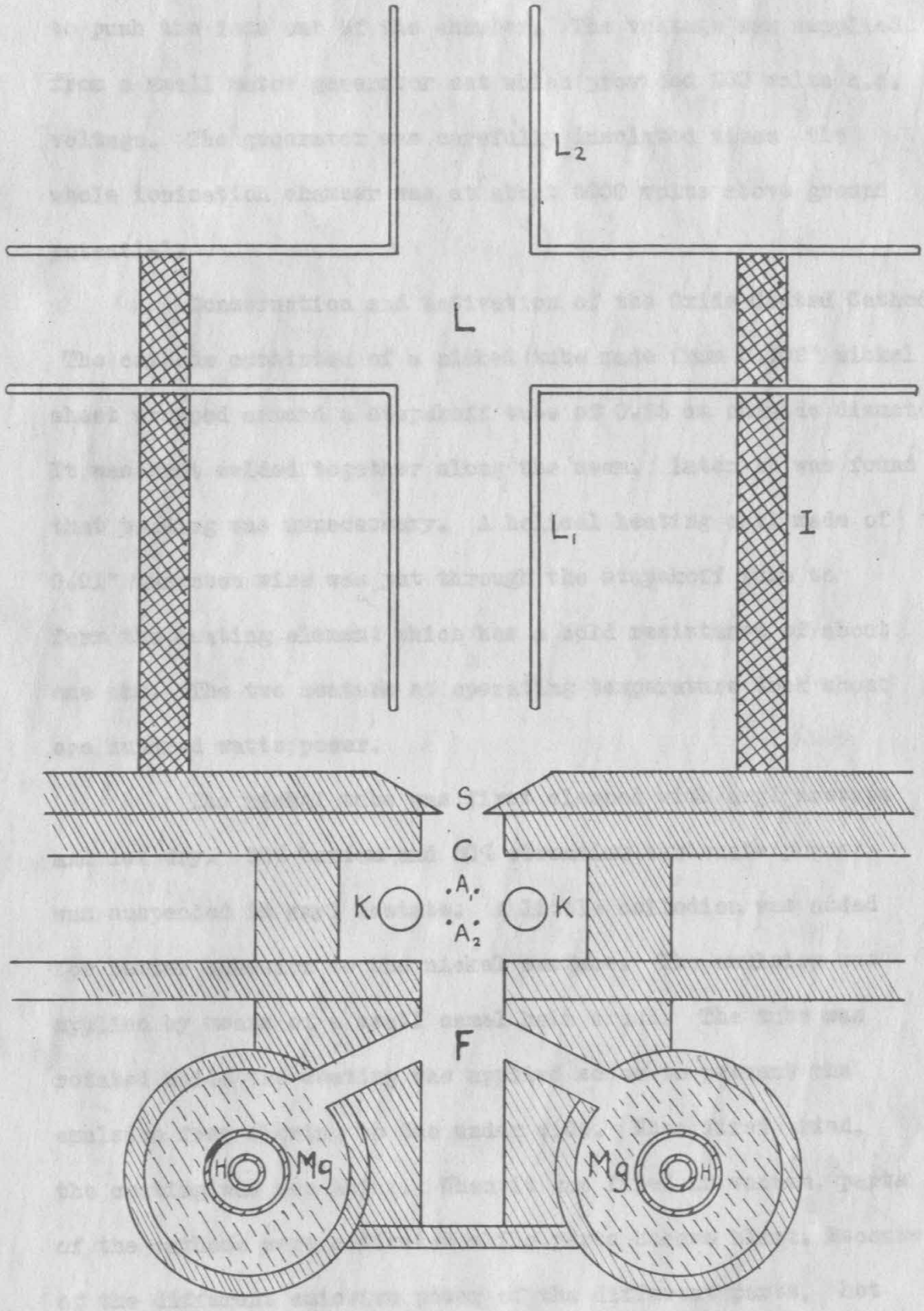
THE ION SOURCE.

The ion source consisted of three parts (see Fig. 5): (1) the furnace F for producing magnesium vapor, (2) the chamber C where the vapor was bombarded by electrons, and (3) the electrostatic lens L by means of which the ions were focussed into a sheet.

(1) The furnace was designed and constructed by J. Evvard. The two thick-walled magnesium tubes were heated in vacuum to supply the vapor. The description can be found in his Ph. D. thesis, 1943.

(2) The ionization chamber C was 1.9 cm wide 0.8 cm deep and 26 cm long. In the middle of the chamber there were four parallel 0.005" tungsten wires held tight by springs. The top two were connected together to form the anode A_1 and the two at the bottom formed the anode A_2. On each side of the four wires, there was one oxide coated cathode also 26 cm long. They were supported by two baked soapstone pieces which closed the two ends of the ionization chamber. The only escape for the ion as well as the magnesium vapor was through the slit S upwards. The two cathodes and the ionization chamber were connected together electrically so that all the electron emission had to go to the two anodes. The anode A_2 was connected to a higher d.c. potential than that of A_1. The electric field thus set up between A_1 and A_2 and that between A_1 and the top of the ionization chamber helped

Magnesium Ion Source



One inch

Fig. 5

to push the ions out of the chamber. The voltage was supplied from a small motor generator set which provided 200 volts d.c. voltage. The generator was carefully insulated since the whole ionization chamber was at about 6000 volts above ground potential.

Construction and Activation of the Oxide Coated Cathode. The cathode consisted of a nickel tube made from 0.002" nickel sheet wrapped around a stupekoff tube of 0.25 cm outside diameter. It was spot welded together along the seam. Later it was found that welding was unnecessary. A helical heating coil made of 0.01" tungsten wire was put through the stupekoff tube to form the heating element which has a cold resistance of about one ohm. The two heaters at operating temperature took about one hundred watts power.

The nickel tube was first cleaned with amyl acetate and let dry. 50% barium and 50% strontium carbonate powder was suspended in amyl acetate. A little collodion was added for better adhesion to the nickel surface. The emulsion was applied by means of a small camel hair brush. The tube was rotated while the coating was applied so as to prevent the emulsion from flowing to the under side. When first tried, the coating was too thick. When it was fired in vacuum, parts of the cathode were activated while parts became black. Because of the different emissive power of the different parts, hot spots were developed along the cathode. This was aggravated

by the fact that the tungsten heater had a larger resistance at the hot section, so more power was dissipated there. At one time, the tungsten heater was burned out at that spot while other parts had not even reached the operating temperature. When the coating was thin, no such uneven heating ever developed.

The whole vacuum chamber was first evacuated to a pressure of about 5×10^{-5} mm Hg. The magnesium heater was started at 2 amperes heating current. When the pressure dropped to the original value, the heating current was increased to 2.7 amperes by small steps and was kept there. If the current was higher than that, the magnesium began to vaporize. The vapor might deposit on the cathode or on the insulator. Both were highly undesirable. When the pressure again dropped to 5×10^{-5} mm Hg., the cathode heater was turned on at 1.5 amperes. As the vacuum improved, the heating current was increased until it reached 2.7 amperes. In no case was the vacuum allowed to go beyond 10^{-4} mm Hg. Now with 100 volts on A_1 and A_2, the emission current of about 40 milliamperes was obtained. This small current was mainly due to space charge limitation. When the spectrograph was actually operating, the cathode heating current was increased to 3 amperes. During the activation period, no voltage on the anodes was necessary. When the chamber was full of ions, the emission went up to as much as 700 milliamperes with only

130 volts on the anodes. By then the four anode wires were red hot.

The above order of turning on the magnesium and cathode heaters was necessary. If the cathode was heated first and the magnesium heater later, the cathode always got contaminated even the vacuum was always better than 7×10^{-5} mm Hg.

(3) The electrostatic lens. Take the center of the lens as the origin of the coordinate system with z-axis along the central straight path of the ions, y-axis perpendicular to the ion beam and the line source, and the x-axis parallel to the line source. If we are not very close to the ends of the lens along the x-direction, the potential function ϕ satisfies the two dimensional Laplace equation

$$\frac{\partial^2 \phi}{\partial z^2} + \frac{\partial^2 \phi}{\partial y^2} = 0 \qquad (1)$$

if we can neglect the space charge effect. This can be solved by means of Schwartz transformation for most of the lenses.

The differential equation [4] for the path of the ion is

$$\frac{d^2y}{dz^2} + \frac{\left[1+\left(\frac{dy}{dz}\right)^2\right]}{2\phi} \frac{\partial \phi}{\partial z} \frac{dy}{dz} - \frac{\left[1+\left(\frac{dy}{dz}\right)^2\right]}{2\phi} \frac{\partial \phi}{\partial y} = 0 \qquad (2)$$

In general, the potential distribution is a very complicated function of y and z. It is practically impossible to get an exact solution of equation (2). However, if we confine ourselves to the discussion of the paraxial ions, we can develope

$\phi(z,y)$ into a power series in y. Then assume

$$\phi(z,y) = \sum_{n=0}^{\infty} \phi_n(z)\, y^n \tag{3}$$

Substituting this into (1) and equating the coefficients of y, we get

$$\phi(z,y) = \sum_{n=0}^{\infty} (-)^n \frac{1}{(2n)!} \frac{d^{2n}\phi_0}{dz^{2n}}\, y^{2n} \tag{4}$$

Therefore

$$\frac{\partial \phi}{\partial z} = \phi_0'(z) - \frac{1}{2!}\phi_0'''(z)\, y^2 + \cdots \tag{5}$$

$$\frac{\partial \phi}{\partial y} = -\phi_0''(z)\, y + \frac{1}{3!}\phi_0^{iv}(z)\, y^3 - \cdots \tag{6}$$

If we only keep the first power of y, equation (2) becomes

$$\frac{d^2y}{dz^2} + \frac{\left[1+\left(\frac{dy}{dz}\right)^2\right]}{2\phi}\phi_0'(z)\frac{dy}{dz} + \frac{\left[1+\left(\frac{dy}{dz}\right)^2\right]}{2\phi}\phi_0''(z)\, y = 0 \tag{7}$$

The paraxial rays never make a large angle with the z-axis, so

$$\frac{dy}{dz} << 1$$

then dropping the subscript for the potential function along the axis, we have

$$\frac{d^2y}{dz^2} + \frac{\phi'(z)}{2\phi}\frac{dy}{dz} + \frac{\phi''(z)}{2\phi}\, y = 0 \tag{8}$$

as the equation of the path of the ion.

In general, even this is still too complicated to integrate. Therefore we have to resort to step-by-step method

of integration. Incidentally, this equation for the path of the ion is independent of e/m, therefore it will be the same for all charged particles. The coefficients of the equation depends on the function ϕ and its derivatives only.

R. Gans [5] gave a simple approximate method of calculating the path. The potential distribution along the lens axis is approximated by a series of straight line segments as shown in Fig. 10. Along each segment

$$\phi''(z) = 0 \quad , \quad \phi'(z) = \text{const.}$$

Equation (8) becomes

$$\frac{d^2y}{dz^2} + \frac{\phi'(z)}{2\phi}\frac{dy}{dz} = 0 \tag{9}$$

Integrated once, we have

$$\frac{dy}{dz}\sqrt{\phi(z)} = C \quad \text{a const.} \tag{10}$$

and twice

$$y = y_0 + \frac{2C\left[\sqrt{\phi} - \sqrt{\phi_0}\right]}{\frac{d\phi}{dz}} \tag{11}$$

where y_0 and ϕ_0 are the respective values of y and ϕ at the beginning of the segment. At the intersection of two segments,

$$\left(\frac{dy}{dz}\right)_2 - \left(\frac{dy}{dz}\right)_1 = \frac{y}{2\phi}\left[\left(\frac{d\phi}{dz}\right)_2 - \left(\frac{d\phi}{dz}\right)_1\right] \tag{12}$$

Equations (10), (11), and (12) are the general formulae for the determination of the ion path and therefore the focal

length of the electrostatic lens.

After finding out that the lens consisting of three parallel plates with slit did not give a well defined beam, it was decided to try another kind as shown in Fig. 6, instead of improving the old one. The reason was that by making the

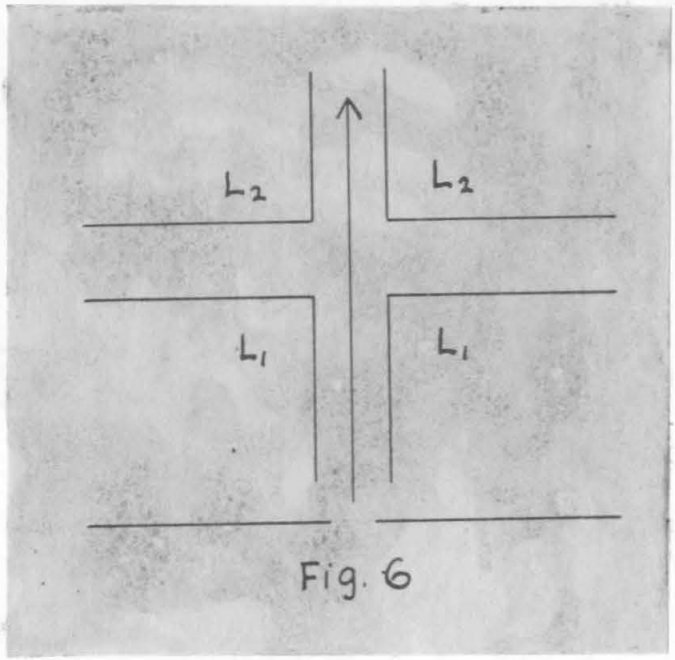


Fig. 6

lower edges of the parallel plates L_1 and L_2 only one half centimeter away from the slit, the field there would be much stronger than that could be obtained with the other type. The potential distribution was solved by Schwartz transformation method in W. R. Smythe, Static and Dynamic Electricity, pp. 294-295. The potential on the axis of the lens which is the z-axis is given by

$$z = \frac{2}{\pi}\left[a \sinh^{-1} \frac{a \tan\theta}{\sqrt{a^2+b^2}} + b \sin^{-1} \frac{b \sin\theta}{\sqrt{a^2+b^2}} \right] \qquad (13)$$

where $\theta = \frac{\pi}{2}\frac{\phi}{V}$ (see Fig. 7). In the present case, $a = b = 1/2$ cm.

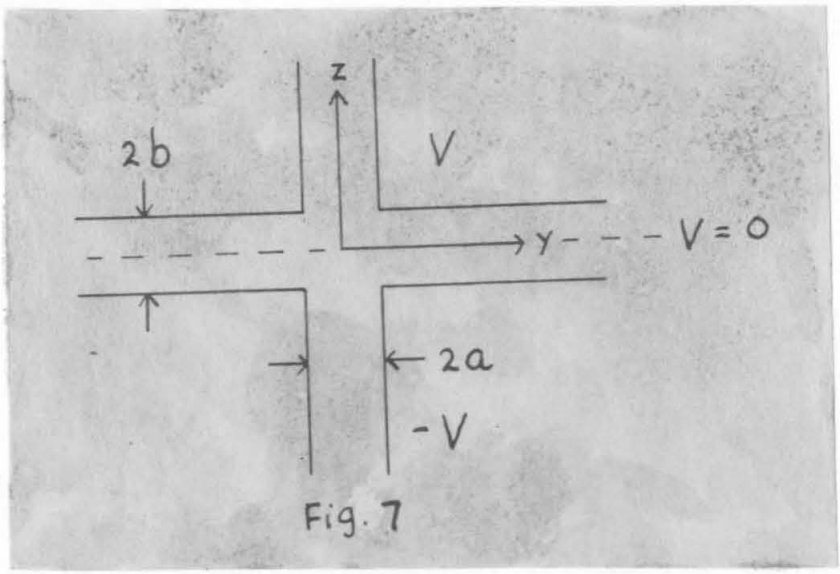


Fig. 7

Therefore

$$z = \frac{1}{\pi}\left[\sinh^{-1}(.707\tan\theta) + \sin^{-1}(.707\sin\theta)\right] \qquad (14)$$

The calculations are listed in Table I. Fig. 8 is a plot of equation (14).

However, in actual use, the two lower plates (Fig. 9)

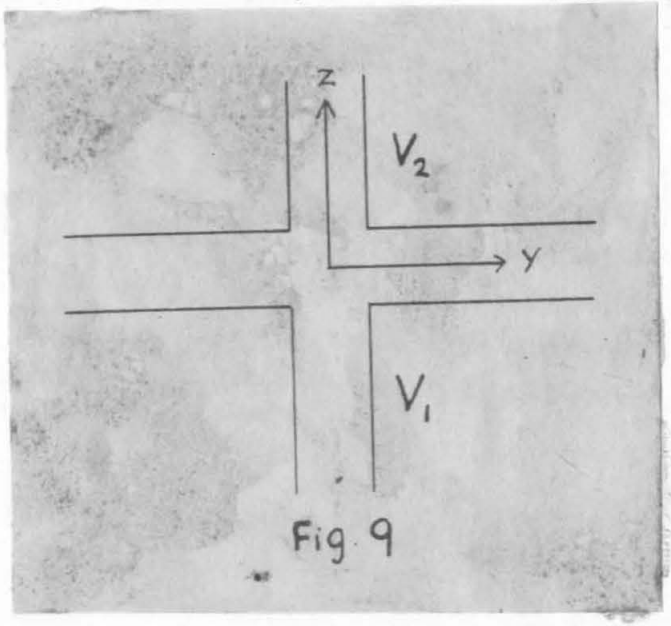


Fig. 9

Potential Distribution Along Axis of Electrostatic Lens

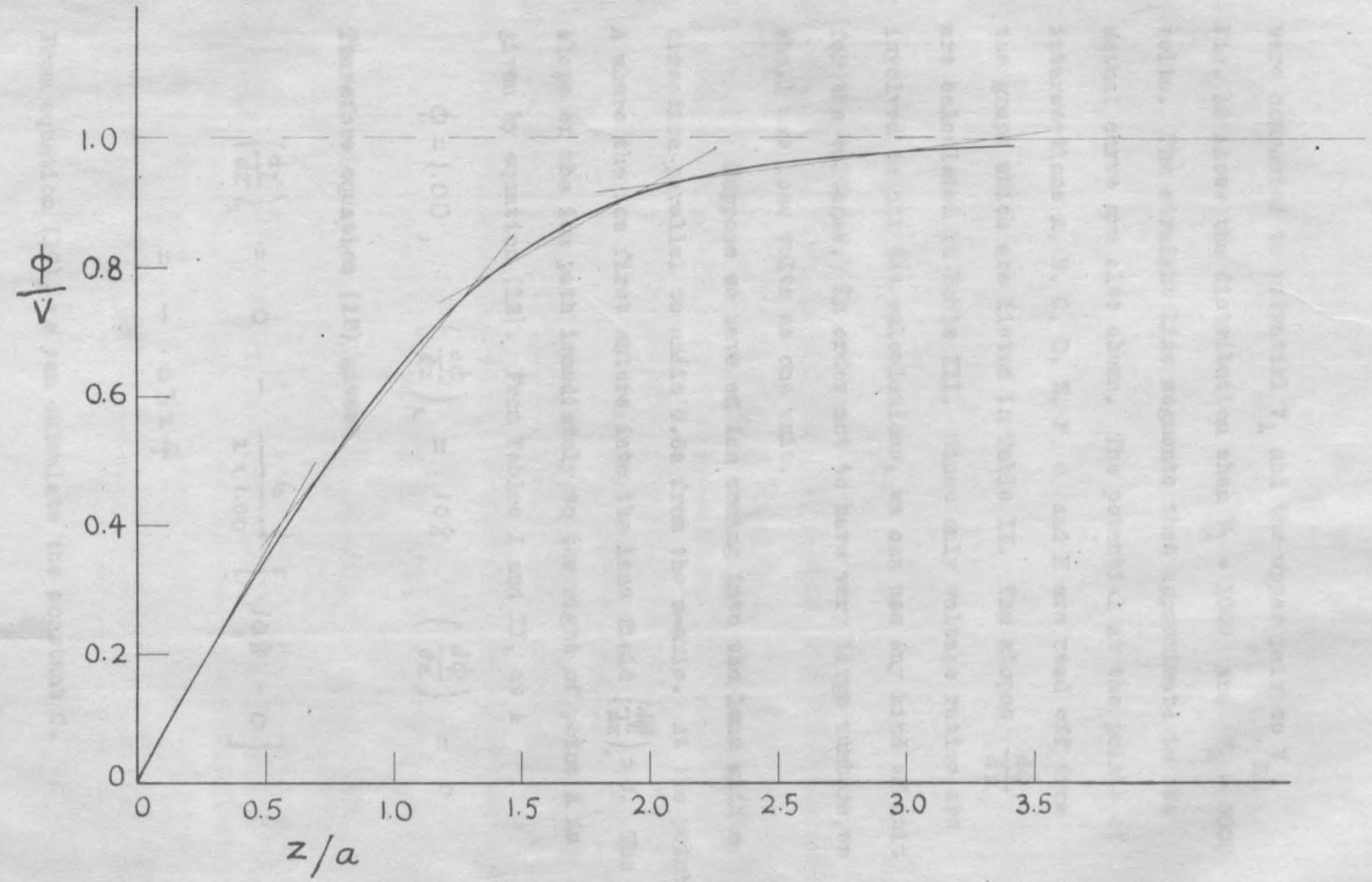


Fig. 8

were connected to potential V_1 and the upper pair to V_2. Fig. 10 shows the distribution when $V_1 = 1000$ and $V_2 = 5000$ volts. The straight line segments that approximate to the actual curve are also shown. The potential at the points of intersections A, B, C, D, E, F, G, and H are read off from the graph which are listed in Table II. The slopes $\frac{d\phi}{dz}$ are calculated in Table III. Since only voltage ratios are involved in all the calculations, we can use any kind of unit for the voltages. In order not to have very large numbers, we shall use looo volts as one unit.

Suppose we have an ion coming into the lens with a direction parallel to and is 0.6a from the z-axis. At the point A where the ion first enters into the lens field $\left(\frac{d\phi}{dz}\right)_1 = 0$. The slope of the ion path immediately to the right of point A is given by equation (12). From Tables I and II, at A

$$\phi = 1.00, \qquad \left(\frac{d\phi}{dz}\right)_2 = .108, \qquad \left(\frac{d\phi}{dz}\right)_1 = 0$$

Therefore equation (12) gives

$$\left(\frac{dy}{dz}\right)_2 = 0 - \frac{.6}{2 \times 1.00}\left[.108 - 0\right]$$

$$= -.0324$$

From equation (10), we can calculate the constant C.

$$C = \sqrt{\phi}\,\frac{dy}{dz} = -\sqrt{1.00} \times .0324 = -.0324$$

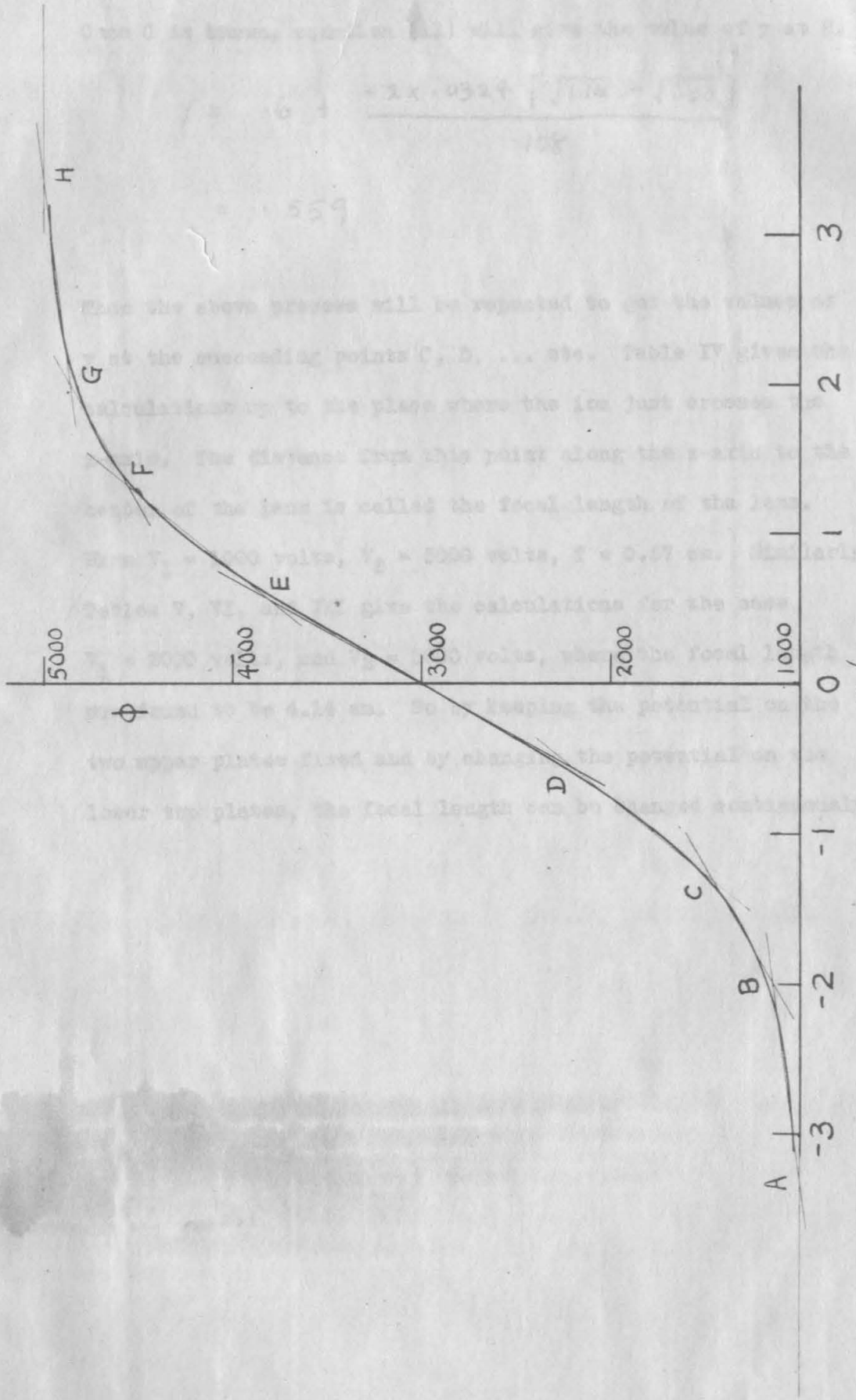


Fig. 10

Once C is known, equation (11) will give the value of y at B.

$$y = .6 + \frac{-2 \times .0324 \left[\sqrt{1.14} - \sqrt{1.00} \right]}{.108}$$

$$= .559$$

Then the above process will be repeated to get the values of y at the succeeding points C, D, ... etc. Table IV gives the calculations up to the place where the ion just crosses the z-axis. The distance from this point along the z-axis to the center of the lens is called the focal length of the lens. When V_1 = 1000 volts, V_2 = 5000 volts, f = 0.67 cm. Similarly Tables V, VI, and VII give the calculations for the case V_1 = 2000 volts, and V_2 = 5000 volts, where the focal length was found to be 4.14 cm. So by keeping the potential on the two upper plates fixed and by changing the potential on the lower two plates, the focal length can be changed continuously.

Table I

$\frac{\phi}{V}$	θ	$.707 \tan\theta$	$.707 \sin\theta$	$\sinh^{-1}$	$\sin^{-1}$	$\frac{z}{a}$
.10	9°	.111	.110	.111	.110	.141
.20	18	.230	.208	.228	.210	.278
.30	27	.360	.321	.368	.327	.442
.40	36	.514	.416	.494	.429	.588
.45	40.5	.604	.460	.572	.478	.668
.50	45	.707	.500	.658	.524	.752
.55	49.5	.828	.538	.754	.568	.840
.60	54	.973	.573	.862	.610	.938
.65	58.5	1.15	.603	.983	.647	1.04
.70	63	1.39	.630	1.13	.681	1.15
.75	67.5	1.71	.654	1.31	.713	1.28
.80	72	2.18	.673	1.52	.738	1.44
.85	76.5	2.94	.688	1.80	.759	1.63
.90	81	4.46	.699	2.20	.774	1.91
.925	83.25	5.98	.702	2.49	.778	2.08
.95	85.5	9.06	.705	2.90	.782	2.34
.975	87.75	18.00	.707	3.58	.785	2.78

Table II

	z	ϕ	$\sqrt{\phi}$	
A	-3.38	1.00	1.00	
				.068
B	-2.08	1.14	1.07	
				.132
C	-1.33	1.44	1.20	
				.276
D	- .58	2.18	1.48	
				.478
E	.58	3.82	1.95	
				.181
F	1.33	4.56	2.14	
				.070
G	2.08	4.86	2.21	
				.031
H	3.38	5.00	2.24	

Table III

	$\frac{d\phi}{dz}$	
∞A	.000	
		.108
AB	.108	
		.292
BC	.400	
		.587
CD	.987	
		.426
DE	1.413	
		-.426
EF	.987	
		-.587
FG	.400	
		-.292
GH	.108	
		-.108
H∞	.000	

Table IV

	y	y_1'	y_2'	C
A	.600	.000	-.0324	-.0324
B	.559	-.0304	-.101	-.108
C	.488	-.090	-.189	-.272
D	.336	-.184	-.217	-.320
E	.120	-.164	-.157	-.307
F	.007	-.144	-.143	-.305

Table V

	z	ϕ	$\sqrt{\phi}$	
A	-3.38	2.00	1.414	
				.039
B	-2.08	2.11	1.453	
				.073
C	-1.33	2.33	1.526	
				.174
D	- .58	2.89	1.700	
				.327
E	.58	4.16	2.027	
				.134
F	1.33	4.67	2.161	
				.053
G	2.08	4.90	2.214	
				.012
H	3.38	5.00	2.236	

Table VI

	$\frac{d\phi}{dz}$	
∞A	.000	
		.085
AB	.085	
		.208
BC	.293	
		.454
CD	.747	
		.303
DE	1.05	-
		-.303
EF	.747	
		-.454
FG	.293	
		-.208
GH	.085	
		-.085
H∞	.000	

Table VII

	y	y_1'	y_2'	C
A	.600	.000	-.0129	-.0180
B	.583	-.0124	-.0411	-.0593
C	.556	-.0398	-.0939	-.143
D	.489	-.0841	-.1097	-.186
E	.383	-.0916	-.0779	-.158
F	.326	-.0731	-.0853	-.126
G	.280	-.0593	-.0534	-.120
H	.252	-.0536	-.0515	

RESULTS

With the above mentioned ionization chamber and electrostatic lens, 120 microamperes were obtained at the collector. The resolution is shown in Fig. 11 with the slit in front of the collector narrowed down to 2 mm.

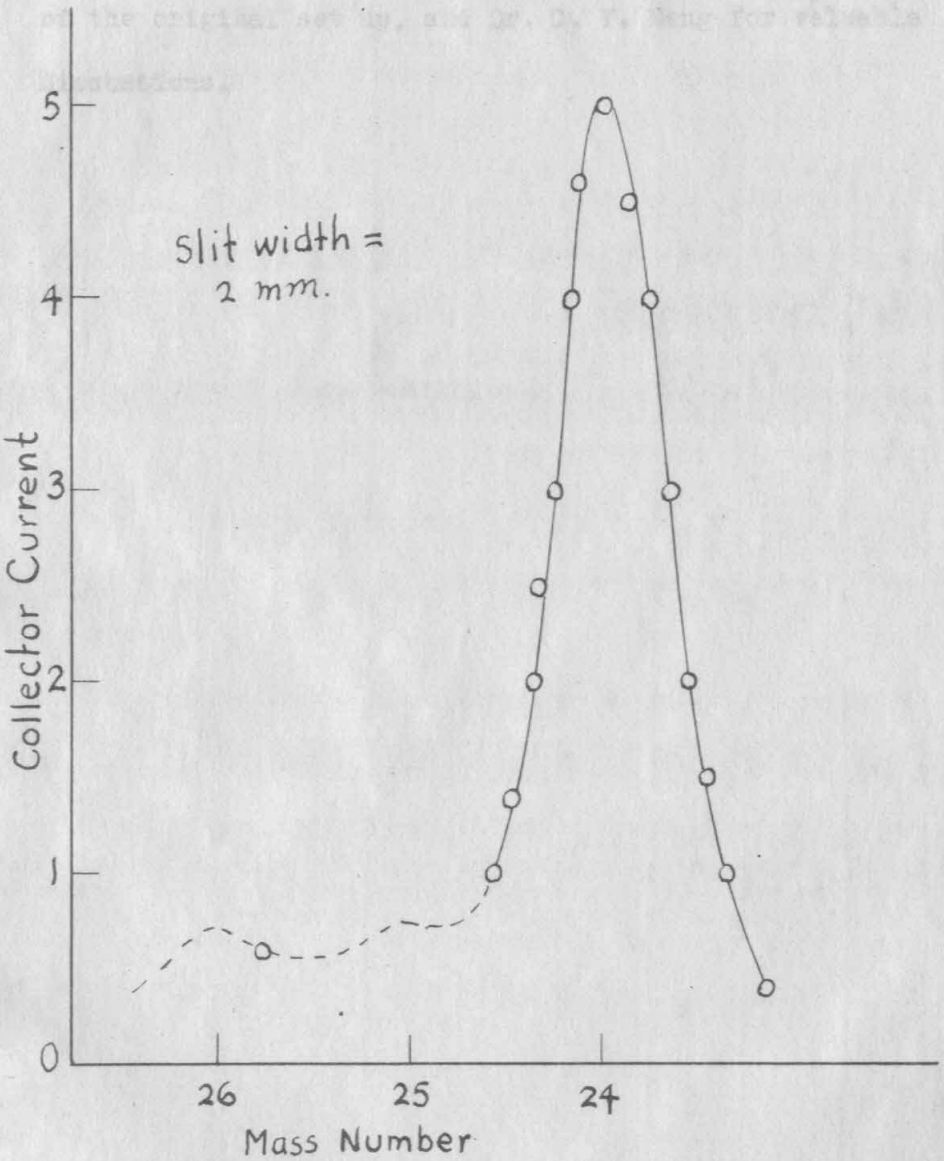

Slit width =
2 mm.
Collector Current
0
1
2
3
4
5
26
25
24
Mass Number

Fig. 11

ACKNOWLEDGMENT.

The writer wishes to express his gratefulness to Prof. W. R. Smythe for his direction and encouragement. He wishes to thank Dr. J. Evvard for showing him the operation of the original set up, and Dr. C. Y. Meng for valuable discussions.

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